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INTERBAND MAGNETOOPTICAL EXPERIMENTS IN $Ga_{1-x}Al_xAs$–GaAs QUANTUM WELLS

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The ground and excited states of two-dimensional excitons originating from different subbands in $Ga_{1-x}Al_xAs$–GaAs quantum wells in a magnetic field are observed using excitation spectroscopy. The exciton binding energies for the different subbands are deduced from the difference in the field dependence of the ground and the excited states as a function of the well thickness. The excited states which are essentially free electron-hole interband transitions are analyzed with theoretical calculations of the Landau levels in a quantum well.

The free excitonic nature of the strong luminescence in good quality GaAs–$Ga_{1-x}Al_xAs$ quantum wells has been demonstrated and investigated for some time [1–4]. One interesting aspect of this phenomenon is that in these quantum wells the well thickness can be made of the order of the exciton Bohr radius. This effect leads to an increase in the exciton binding energy with decreasing thickness, because the exciton will behave progressively as a quasi two dimensional hydrogen atom. This trend has been shown theoretically by several authors [2, 5, 6], and experimentally some indication of this effect has been inferred from the thickness dependence of the difference between the ground and the first excited state of the exciton [2]. In a magnetic field the exciton continuum splits into discrete excited states which are weakly bound to the Landau levels and which can therefore be described as free electron and hole states. The lowest bound excitonic states on the contrary experiences only a comparatively weak diamagnetic shift. On the one hand, the simultaneous observation of the magnetic field dependence of the bound and the continuum states allows a direct determination of the band edge and the exciton ground state energies and thereby of the binding energy in the quantum well. On the other hand electron and hole Landau levels in a quantum well can be investigated by looking at the field dependence of the exciton continuum states. There has recently been an increased interest both theoretically [7, 8] and experimentally [9, 10] in hole Landau levels in quantum wells.

Four samples have been studied with different GaAs layer thickness and Al content (See table I). The intensity of the luminescence of the lowest energy transition (heavy-hole exciton ground state) is measured as a function of the excitation intensity at different fixed values of the magnetic field. Both the incident and the emitted radiation were at right angles to the layer plane and parallel to the magnetic field axis (Faraday configuration). The exciting light was left or right circularly polarized with respect to the magnetic field. Magnetic fields up to 23 T were made with polyhelix resistive magnet. The radiation with wavelengths between 804 and 740 nm was generated with Kr laser pumped CR-599 dye laser with LD700 as a dye.

In fig. 1 the excitation spectra for one circular polarization are shown for different values of the magnetic field. In this case the spectrometer was placed on the center of the luminescence line and the excitation spectra of the higher lying transitions are observed. The two lower lying transitions which partly coincide with the liminescence line and which are due to the two exciton ground states associated with the two lowest hole subbands were measured separately with the spectrometer placed in the low energy

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Table I
Sample parameters and experimentally determined masses

Sample thickness (nm)	Al content	Reduced masses E–HH	Reduced masses E–LH	Hole masses HH	Hole masses LH
5	0.18	0.084	—	>1	—
9	0.29	0.079	—	>1	—
10	0.29	0.077	0.057	>1	0.2 ± 0.1
12.5	0.21	0.069	0.064	$0.7+0.2$	0.35 ± 0.1

tail of the luminescence. The spectra which are featureless at zero magnetic field in this energy range show a very rich structure in a magnetic field as a consequence of the development of Landau levels. Fig. 2 shows a plot of the transition energies vs. magnetic field for the two different polarization directions. The results shown are for the 12.5 nm thick quantum well. To bring some order in the experimental results lines are

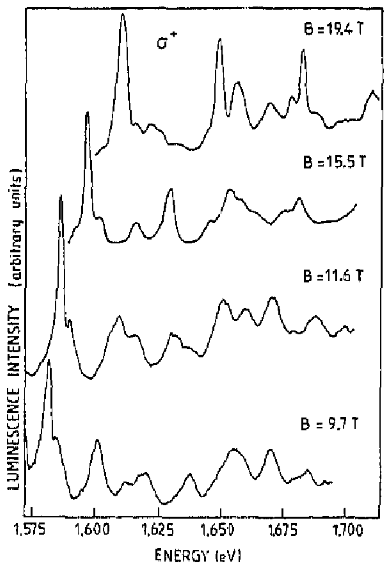
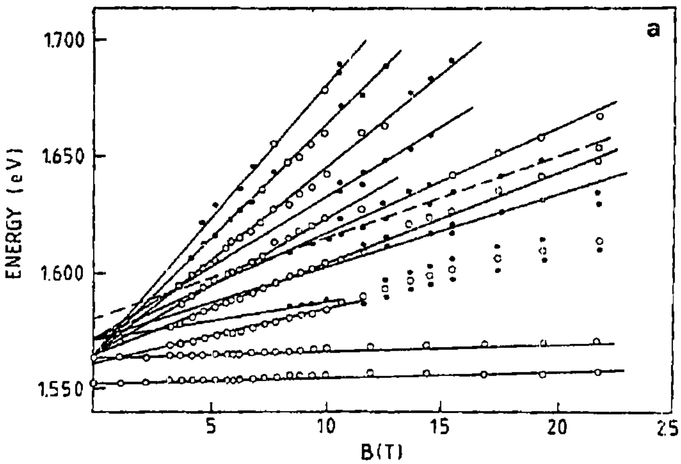


Fig. 1. Excitation spectrum at different values of the magnetic field for the 12.5 nm sample. The spectrometer is placed at the center of the luminescence line and the higher lying transitions are measured.

drawn to connect the observed maxima in fig. 2. This way one notices that the two transitions at the lowest energies show only a very weak magnetic field dependence; that the next transition at higher energy shows a stronger magnetic field dependence and extrapolates approximately to the same energy as the second transition at zero magnetic field. Furthermore, at higher energies the transitions can be distinguished in two sets, each set extrapolating to the same energy at zero magnetic field, but to a different energy for both sets. The two transitions at low energies are interpreted as the exciton ground state for the heavy hole (HH) and the light hole (LH) exciton. These ground states are only weakly affected by the magnetic field since the Coulomb energy dominates over the magnetic energy. The higher excited exciton states are much more weakly bound and in this case the magnetic field determines the energy spectrum, as is clear from the fact that the curves extrapolate to the same energy at zero magnetic field. Therefore, the two sets of transitions extrapolating to the same energies can be analyzed as free electron and hole band to band transitions. The transition which appears like a splitting of the LH exciton ground state is interpreted as a first excited excitonic state of the HH which for this sample is accidentally degenerate with LH exciton ground state. It is indeed observed in the thinner samples, where the splitting between the hole subbands is larger, that this transition does not extrapolate to the same energy at zero magnetic field as the LH ground state. Not all observed transitions have such a simple magnetic field dependence. As is clear from fig. 2 the strength of some of the transitions is dependent on the



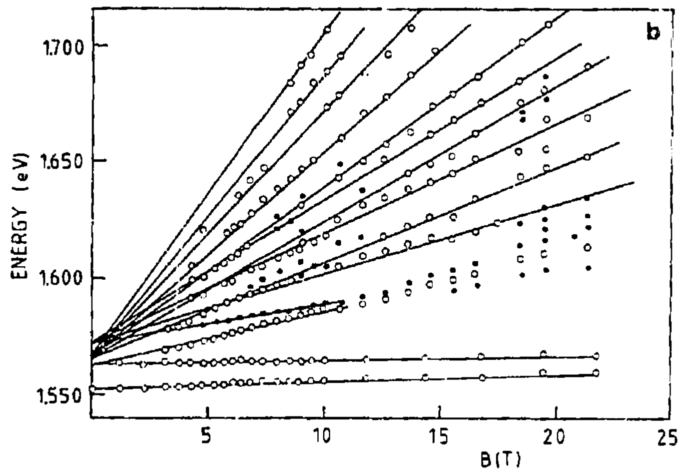


Fig. 2. Energy of the maxima in the excitation spectra as a function of magnetic field for σ^+ (a) and σ^- (b) polarization of the exciting light. The open circles correspond to strong and the full circles to weak transitions. The drawn lines are a guide to the eye.

magnetic field up to the point that some of them even become unobservable at higher fields. In addition there exist several weaker peaks which cannot be assigned to a one of the sets described above.

We determine the exciton binding energy directly from the spectra as the difference in energy between the ground state and the continuum states at zero magnetic field as obtained from the extrapolation of the field dependence of

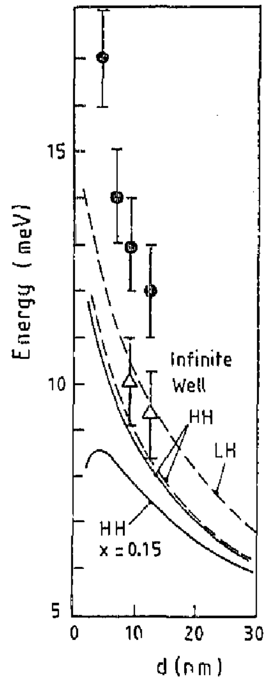


Fig. 3. Exciton binding energy for the HH exciton (closed circles) and the LH exciton (triangles) as determined from magnetic field dependence of the excitation spectra. The lines represent theoretical calculations of the thickness dependence of the binding energy for an infinite well by Miller et al. [2] (dashed lines) and for infinite and finite height wells by Greene et al. [3] (solid lines).

the excited states. In fig. 3 the binding energies obtained in this fashion as a function of the well thickness are shown. Also included in the figure are the results of theoretical calculations by Greene et al. [6] and by Miller et al. [2]. It can be seen that both experimentally and theoretically an increase in the binding energy is found with decreasing layer thickness. However, the experimental values are much higher than the theoretical ones. In addition the so called LH exciton is found to be more weakly bound than the HH exciton, which is contrary to the theoretical predictions.

As can be seen from fig. 2 several Landau level like transitions are observed for both subbands. Following refs. 7 and 8, we have calculated the Landau levels for the holes and the electrons in a quantum well. In this case each material is represented with a six-band model i.e. a spin up and a spin down conduction band and the fourfold $J=3/2$ valence band set, split into heavy-hole and light-hole bands at $k \neq 0$. The theory described in refs. 7 and 8 was developed for the case of a superlattice, however since the coupling between GaAs wells becomes negligible for sufficiently thick $Ga_{1-x}Al_x$-As layers the results can be directly applied to the case of quantum wells. In a magnetic field the envelope wavefunction for the motion along the plane is described by a six-component vector,

$$\begin{aligned}\Psi_n = (&c_1(z)\varphi_n, c_2(z)\varphi_{n-1}, c_3(z)\varphi_{n+1},\\ &c_4(z)\varphi_{n+1}, c_5(z)\varphi_n, c_6(z)\varphi_{n+2})\end{aligned} \quad (1)$$

where φ_n are harmonic oscillator functions, $n = -2, -1, 0, 1, \ldots$ and $c_i(z)$ (z is the direction of the magnetic field and the superlattice growth direction) coefficients which are automatically vanishing for those components which have a negative oscillator index. The six components of the envelope wavefunction correspond to the CB↑; $J_z=3/2$; $J_z=-1/2$; CB↓; $J_z=+1/2$ and the $J_z=-3/2$ cell periodic parts of the Bloch wavefunction. The coefficients c_i are determined from the boundary conditions imposed on the wavefunction (1) of the two materials at the interface. These boundary conditions are derived from the continuity of the current operator at the interface for a sample with a given thickness and Al content and at a given magnetic field. These coefficients are therefore dependent on the sample parameters and on the magnetic field. The wavefunction (1) implies a selection rule of $\Delta n = \pm 1$ for interband transitions. In fig. 4 the calculated Landau levels and the allowed transitions for the sample with 12.5 nm thickness are shown. A striking feature of this figure is the complex, strongly non-linear behaviour of the hole Landau levels. This non-linearity comes from the fact that at $k_\parallel = 0$, or equivalently, at

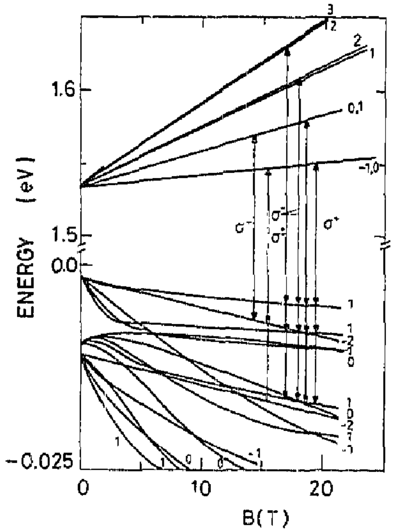


Fig. 4. Calculated Landau levels for the conduction and for three hole subbands of a 12.5 nm GaAs–$Ga_{0.79}Al_{0.21}$ quantum well. The arrows indicate allowed transitions for σ^+ and σ^- radiation ($\Delta n = +1$ or -1). The numbers correspond to the Landau level quantum numbers.

$B = 0$ heavy and light hole subbands are decoupled. At finite values of the magnetic field Landau levels originating from the first hole subband interact with Landau levels from the next hole subband and gives rise to the anticrossing behaviour as can be seen in fig. 4.

We have tried to compare the calculated energy differences for allowed transitions with the experiments using the $\Delta n = +1$ or -1 selection rule with the corresponding experimental results for left and right circular polarization (figs. 2a and b) and have found that we can explain the experiments in this way in the sense that there exist calculated transition energies vs. magnetic field which correspond to the experimental observations. However using the calculated energy levels and using only the selection rule $\Delta n = \pm 1$, much more transitions are predicted than that are experimentally observed. Furthermore, this simple utilisation of the calculated Landau levels does not permit to calculate the strength of the transition, whereas it is clear from fig. 1 that different transitions have quite different strengths. The conclusion is therefore that in order to make a realistic comparison between theory and experiment the overlap matrix elements of the envelope wavefunction of the initial and final states must be calculated. In the case of simple non-interacting bands the matrix elements can easily be evaluated in terms of the parity (even or odd) of the wavefunction. Due to the interaction between the different Landau levels this is not possible in the present case. In addition the coefficients c_i in (1) are strongly field dependent for the same reason which implies that the transition strength is also field dependent. Qualitatively such a behaviour can indeed be observed in fig. 2 where the same transition may change its strength for different magnetic fields.

As a consequence of the difficulties encountered with the comparison of the theory with the experiments we have analysed the data in a more simple manner. From a plot of the slopes of the different transitions as a function of the Landau level quantum number a straight line is found within experimental error. From the slope of this line a reduced mass for the combined electron and hole Landau level splitting can be determined and the results are given in table 1. It is clear from this table that the reduced masses for the transitions involving the heavy hole subband are almost equal to the electron mass alone taking nonparabolicity into account. This implies that the heavy hole mass is much larger than the electron mass. Transitions involving the next hole subband, which can only be observed clearly in two samples show a reduced mass which is definitely lighter than the electron mass and a hole mass of this subband can be determined. Qualitatively these results are in agreement with the theoretical expectations. It is clear from fig. 4 that as a consequence of the anticrossing the highest hole Landau levels have a very weak field dependence whereas the set of Landau levels originating from the next band are much more strongly field dependent. (Note in fig.

4 that the energy scale of the holes is five times as large as that of the electrons.)

The experimentally determined reduced masses are strikingly different from those used in the theories to calculate the exciton binding energy, which were $0.04m_0$ for the heavy hole and $0.051m_0$ for the light hole exciton. These values are based on a simplified description of the valence subband dispersion which neglects the coupling between the bands. We find that not only the masses are heavier than expected but also that the heavy hole–electron reduced mass is much heavier than the light hole–electron reduced mass. This difference arises from the fact that in the theories the coupling between the different hole bands has been neglected, whereas it is clear from the calculation of the Landau levels in fig. 4 that inclusion of this coupling indeed leads to an increase of the masses. The discrepancy between the calculated and measured binding energies in fig. 2 arises in fact from the difference in the experimentally determined reduced masses and those used in the theories. Since in a hydrogenic model the binding energy is proportional to the reduced mass we can scale the theoretical curves for the binding energies with a factor which is the ratio of the experimental value of the reduced mass to the theoretical value. This way the lowest two exciton ground state energies can be made to agree with the theory within experimental error.

In summary it may be stated that we have observed the ground state and several excited states of the quasi two dimensional exciton in GaAs–$Ga_{1-x}Al_xAs$ quantum wells as function of magnetic field with excitation spectroscopy. The exciton binding energies as a function of thickness is obtained from this data using the different magnetic field dependence of the ground and the excited states. The experimentally determined binding energies are enhanced with respect to the bulk values and increase with decreasing thickness. The experimental values obtained however are higher than theoretical predictions [2, 6]. This discrepancy disappears when the experimentally determined reduced masses for the different subbands are used instead of the value used in the theories. Furthermore, we have compared the magnetic field dependence of the higher free carrier like excited states with calculations of the Landau levels in a quantum well including the interaction between the different hole subbands. It is found that the theory predicts more transitions than actually observed if one imposes a selection rule based on the cell periodic part of the wavefunction only. For a detailed comparison the full overlap matrix elements should be calculated. Qualitatively the magnetic field dependence of the theoretically calculated Landau levels of the different subbands agree with that observed experimentally in the sense that both theoretically and experimentally a very heavy hole mass is found for the highest subband and a much lighter hole mass for the second subband. The high value of the masses involved in the hole subbands which is observed experimentally both in the value of the exciton ground state energies as in the field dependence of the excited states is a consequence of the coupling between light and heavy hole subbands at finite k-values which has been neglected in the calculations of the binding energies. As such our results provide experimental evidence for this effect.

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